RADIUM AND RADIUM DECAY PRODUCTS IN SOME MACROMYCETES: FIRST RESULTS

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ABSTRACT - First results of measures of radium and radium decay products on some species of fungi are presented here. These fungi could not really be used as bio-indicators, because in most cases the transfer factor was inferior to one, but the health problem which might arise in some cases is underlined. The fungi grew either in areas polluted by the uranium industry or in other areas with very diversified biotopes. Apart from the radioactivity, measures were made on stable lead and on the pH of the soils.

RÉSUMÉ - Les premiers résultats de mesures de radium et de descendants du radium sur un certain nombre d'espèces de champignons sont présentés ici. Ces champignons ne peuvent pas réellement être utilisés comme bio-indicateurs, le facteur de transfert sol/carpophore étant généralement inférieur à 1, mais le problème sanitaire potentiel est souligné. Les champignons poussaient soit dans des zones polluées par l'industrie de l'uranium, soit dans d'autres zones avec des biotopes très divers. Des dosages de plomb stable ont été réalisés dans certains cas et le pH des substrats a été mesuré.

KEY WORDS : Natural radioactivity, Radium, Lead, Transfer factors, Uranium industry.

INTRODUCTION

Following the Chernobyl accident, many scientists studied the presence of fission products - in particular cesium isotopes - in fungi. The only natural radionuclide which was taken into consideration was potassium (K 40) (Elstner et al., 1987; de Meijer et al., 1988), because of its chemical affinity with cesium. However there are numerous natural radioelements in the soil and human activities may concentrate them and make them more available for living organisms. Some of these radionuclides are very radiotoxic and their migration should be followed with care. First results of absorption of radium 226 and some of its daughter products by some species of fungi growing on specific and differently contaminated soils are presented here. Instead of adopting a statistical approach, which does not provide many informations about transfer mechanisms, a case by case study was realised, as has also be done in some studies on radioactive cesium (de Meijer et al., 1988): each result represents only one sample, but it is completed by the description and analysis of different parameters:

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Source : MNHN, Paris

general ecology, measurements of radioactivity and stable lead in the soil as well as in the fruitbodies.

METHODS

The samples

Coprinus comatus (Müll: Fr.) Pers N° 1 and *Coprinus atramentarius* (Bull.: Fr.) Fr. N° 1 were collected just a few meters from the fence surrounding a tip of residues of a yellow cake plant in Gueugnon (France). This place is now urban wasteland partly rehabilitated as leisure area and was supposed to be very rich in all uranium decay products. *Hypholoma fasciculare* (Huds.: Fr.) Kumm., was collected on a dead branch burried on the tip itself. Its direct substrate, i.e. the branch, was not analysed, but the earth at the exit of a rabbit burrow a few meters away was.

Agaricus campestris L.: Fr. and *Macrolepiota excoriata* (Sch.: Fr.) Wasser, were collected in a meadow, about 100 meters from a repository for long lived nuclear waste (officially only thorium 232 and radium 226) in Issy l'Evêque (France). Before that, the site was a uranium mine and supposed to be rich in uranium and all its decay products.

Leccinum versipelle (Fr. & Hök.) Snell and *Coprinus atramentarius* N° 2 were collected on a slag heap called "terril de l'Héribus" in Mons, Belgium. At present it is mainly colonised by birches. *Xerocomus chrysenteron* (Bull.) Quél. and *Xerocomus badius* (Fr.) Kühn. ex E.J. Gilbert, were collected in a coniferous forest in Czechoslovakia, about 40 kilometers north of Brno; these were originally intended for the monitoring of cesium from Chernobyl.

Hydnum repandum L.: Fr. and *Coprinus comatus* N°2 were collected in deciduous forests near Cluny, in Burgundy, France.

Except *X. badius* all fruitbodies (between 8 and 15) of each sample were taken on the same spot and had to be sufficient to fill at least a Petri plate format container after having been dehydrated and crushed.

For the two *Coprinus* of Gueugnon and for the *Coprinus comatus* of Cluny, two samples of soil were analysed: one taken under the fungi, the other about three meters away. In the case of *Coprinus comatus* of Gueugnon, we indicate both figures, the first corresponding to the soil directly under the fungus. In the other cases, there were no relevant differences. For these three samples, preliminary results had already been published (Daillant, 1991), but for *Coprinus atramentarius* and *Coprinus comatus* from Cluny, the samples were measured again to try to reduce the margin of error.

We report also the results of a Pb 210 measure on another *C. comatus* from Saint Denis de l'Hôtel (Loiret, France).

The species of *Coprinus, Hypholoma fasciculare, Agaricus campestris* and *Macrolepiota excoriata* are considered saprophytic fungi; *Leccinum versipelle, Hydnum repandum, Xerocomus badius* and *Xerocomus chrysenteron* are mycorrhizal.

Radionuclides examined

The radionuclides examined here are all of the family of uranium 238, which is one of the three natural radioactive families. It is presented on table 1.

Source : MNHN, Paris

Uranium 238 (4,5x10 E9 a) → Thorium 234 (24,1 d) → Protactinium 234 (1,2 m) → Uranium 234 (2,5x10 E5 a) → Thorium 230 (8x10 E4 a) → Radium 226 (1600 a)
↓
Radon 222 (3,8 d)
↓
Polonium 210 (138,4 d) ← Bismuch 210 (5 d) ← Lead 210 (22 a) ← Polonium 214 (162 µs) ← Bismuch 214 (19,8 m) ← Lead 214 (26,8 m) ← Polonium 218 (3 m)
↓
Lead 206 (stable)

Table 1: Uranium 238, its decay products and their half-lives (only the main disintegrations are shown here); the letters stand for the following periods: a: Year, d: Day, m: Minute, s: Second.

Tableau 1: La famille de l'uranium 238, avec ses produits de filiation et leurs périodes (seules les désintégrations principales figurent sur ce tableau); les lettres indiquent les périodes suivantes: a: année, d: jour, m: minute, s: seconde.

The measurements were made in gamma-spectrometry: the apparatus used were 3 hyper-pure germanium detectors of 2 types (1 Canberra type P, resolution 1,7 KeV at 1,33 MeV and an efficiency of 13% and one Ortec, type N with the same resolution and an efficiency of 24%).

Because of the method used, only the nuclides emitting gamma photons at sufficiently high energies could be measured. Furthermore, the short-lived radon decay products cannot be considered separately from radium. Thus the radionuclides examined here are radium 226 and, in some cases, lead 210. Thorium 234 was also measured but the results are not presented here.

Radium was either measured directly, using its 185 KeV line or indirectly using the lines of its daughter products:

- It was measured directly for the two species of *Coprinus* from Gueugnon and the soil on which they were growing; however, the 185 KeV line is common to radium 226 and uranium 235, so the latter has to be evaluated (according to the natural proportions) and substracted; this explains partly the high margins of error.

- It is therefore preferable to measure radium 226 using its decay products, i.e. bismuth 214 (609 KeV) and lead 214 (352 and 295 KeV), at least in some cases. This was done for the other samples; to do so, it is necessary to allow the decay products to reach equilibrium with radium 226; its first decay product is radon 222, a gas: it diffuses easily. After dehydration at 105°, the samples were therefore sealed in a tight container for at least four periods of radon 222, reaching thus 95% equilibrium between radium and the short lived decay products Pb 214 and Bi 214.

The results of the Ra 226 measures are on table 2: if measured directly, only the figure is indicated, expressed in becquerel per kilo dry matter. If measured on the line of bismuth 214, the symbol Bi is put into brackets after the figure. If measured on the line of lead 214, the symbol Pb is put into brackets after the second figure. Bismuth was used first and lead is only indicated if it can reduce the margin of error: eg. *Agaricus campestris* of Issy l'Evêque: although the margin of error for Bi 214 might

Source : MNHN, Paris

lead to a result as low as 15 Bq/Kg, the result for Pb 214 is: 49+/-23 which means that the corresponding radium content is not inferior to 26 Bq/Kg and is written on the table as: 37 +/-22 (Bi)
49 +/-23 (Pb)

Lead 210 was measured on its 46 KeV line; the results are reported on table 3.

The count time ranged from 4 to 24 hours.

Total (inert) lead was also measured chemically on three samples of *Coprinus* and the soil on which they grew: the digestion of samples was done at the University of Mons, department of professor Piérart and the analysis (atomic absorption) was done at the ARGUK laboratory, Oberursel (FRG), Dr Maraun.

The pH of the soil was also measured, using a conventional glass electrode.

Site	species	pH	Ra 226 in soil	Ra 226 in fungus	concentration
Gueugnon	C. comatus	3,7	216 +/- 30	136 +/- 87	0,6
			and 157 +/- 27		-
	C. atramentarius	sim.	245 +/- 35	135 +/- 110	0,5
	H. fasciculare	*	*	< 18 (Bi)	-
Mons	L. versipelle	5,8	56 +/- 9 (Bi)	< 6,4 (Bi)	-
	C. atramentarius	id.	68 +/- 18 (Bi)	< 17 (Bi); < 7 (Pb)	-
Cluny	C. comatus	7,7	67 +/- 10 (Bi)	66 +/- 72	1
	H. repandum	4,5	43 +/- 15 (Bi)	32 +/- 14 (Bi)	0,7
Czechoslovakia	X. chrysenteron	3,6	76 +/- 13 (Bi)	39 +/- 22 (Bi)	0,5
	X. badius	sim.	sim.	22 +/- 10 (Bi)	0,3
Issy l'Evêque	A. campestris	5,3	124 +/-35 (Bi)	37 +/- 22 (Bi)	0,3
				49 +/- 23 (Pb)	0,4
	M. excoriata	id.	id.	< 29 (Bi); < 26 (Pb)	-

Table 2 : Results of radium measurements in the soils and in the fungi, expressed in Bq/Kg (dry matter) with the concentration factor and the pH of the soil. The abbreviation sim. means that the nature of the sample is similar to the preceding; the abbreviation id. means that the nature of the sample is identical to the preceding.
**H. fasciculare* does not grow on the ground; the soil a few meters away was nevertheless analysed and showed a contamination of 11 300 Bq/Kg of radium 226. The chemical symbols in brackets indicate whether radium was measured on the line of bismuth 214 (Bi) or on the lines of lead 214 (Pb). If measured directly on the line of radium, nothing is added to the figure. In the case of *C. comatus* of Gueugnon, we indicate the two results, the second corresponding to the soil 3 meters away of the fruitbodies.

Tableau 2: Résultats des mesures de radium dans les sols et les carpophores, exprimés en Bq/Kg (matière sèche) avec le facteur de concentration et le pH du sol. L'abréviation sim. signifie que la nature de l'échantillon est similaire au précédent; id. signifie que l'échantillon est identique au précédent.
**H. fasciculare* ne pousse pas sur le sol; un échantillon de sol à quelques mètres des carpophores a néanmoins été analysé et présentait une contamination en radium 226 de 11 300 Bq/Kg. Les symboles chimiques entre parenthèses indiquent si le radium a été mesuré à partir des pics de bismuth 214 (Bi) ou du plomb 214 (Pb). S'il a été mesuré directement, aucune indication ne figure après le chiffre. Dans le cas du substrat de *C. comatus* de Gueugnon, nous indiquons deux résultats, le second correspondant à l'échantillon de sol prélevé à 3 mètres des carpophores.

Source : MNHN, Paris

Site	species	pH	Pb 210 in soil	Pb 210 in fungus	concentration
Gueugnon	C. comatus	3,7	106 +/- 17	722 +/- 142	6,8
		sim.	and 54 +/- 14		-
Czechoslovakia	X. chrysenteron	3,6	< 29	387 +/- 179	>13
	X. badius	sim.	sim.	< 60	-
Cluny	C. comatus	7,7	70 +/- 14	61 +/- 73	0,9
	H. repandum	4,5	53	< 78	-
St. Denis	C. comatus	NR	NR	134 +/- 83	-

Table 3: Results of measurements of lead 210 in some fungi and the corresponding soils, expressed in Bq/Kg (dry matter) with the pH of the soil; the concentration factor is only indicated when the results allow it. As for radium, two results are mentioned for Gueugnon. (sim. means that the sample is similar to the preceding. NR stands for not recorded).

Tableau 3: Résultats des mesures de plomb 210 sur quelques carpophores et les sols correspondants, exprimés en Bq/Kg (matière sèche) avec le pH du sol; le facteur de concentration n'est indiqué que si les résultats le permettent. Comme pour le radium, deux résultats sont fournis pour le substrat de *C. comatus* de Gueugnon. (sim. signifie que l'échantillon est similaire au précédent. NR signifie non relevé).

RESULTS

The results for 226 Ra are on table 2. The concentration factor is never superior to 1, whether the fungi are saprophytic or mycorrhizal. In non polluted areas, the order of magnitude is of some tens of becquerel per kilo dry matter. Table 3 shows the results for 210 Pb, with a very high concentration factor in two cases and concentrations of hundreds of Bq/Kg. The total lead measures are on table 4 : the highest concentration factor is 0,17.

DISCUSSION

The first comment which has to be made is that the margins of error are fairly high, this is partly due to the measuring procedure which was mentioned in the methods, but there are other difficulties: the precision is proportional to the quantity of sample material and to the count time.

The quantity of soil is not limited, but it is often very difficult to find enough fungi of the same species on the same spot: the measurement procedure being much more difficult as for - say - cesium 137 and cesium 134 measurements, a larger quantity had to be gathered: as for cesium 300 g of fresh matter are ideal and 30 or 40 g may be sufficient if the count time is long enough, the ideal quantity of fresh sample material is of at least 500 g for radium and its decay products. As an example, the spectrum of *Xerocomus chrysenteron* (Fig. 1) with the main lines and the background gives an idea of the added difficulty.

The results are nevertheless relatively concordant and make some first conclusions possible even though the number of samples is still limited.

Source : MNHN, Paris

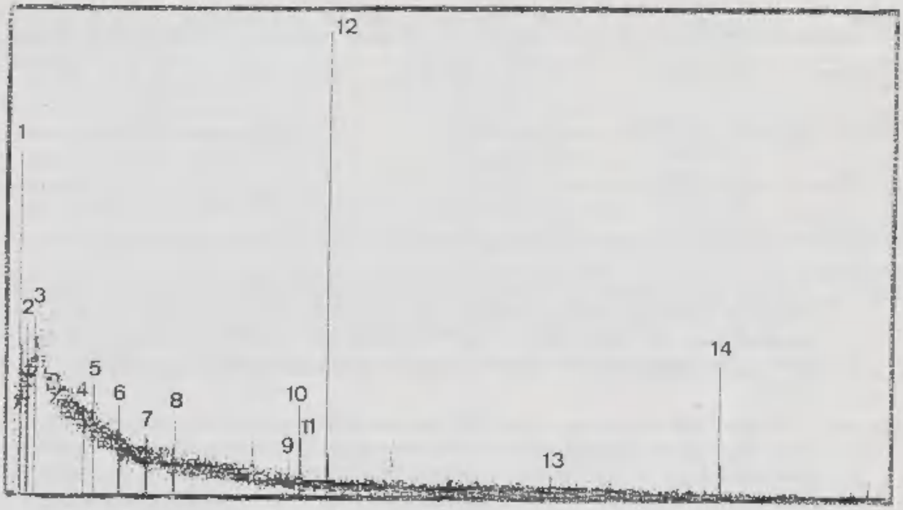

Figure 1: Spectrum of *Xerocomus chrysenteron* (Czechoslovakia). One can clearly see the background; the elements and the energies of the main lines are indicated below: 1) x emission of barium (decay product of cesium 137); not used for measurements. 2) Lead 210 (46 KeV) 3) Thorium 234 (63 KeV) 4) Uranium 235 (163 KeV) 5) Radium 226 and uranium 235 (186 KeV) 6) Lead 212 (238 KeV) 7) Lead 214 (295 KeV) 8) Lead 214 (352 KeV) 9) Thorium 208 (583 KeV) 10) Cesium 134 (604 KeV) 11) Bismuth 214 (609 KeV) 12) Cesium 137 (661 KeV) 13) Bismuth 214 (1120 KeV) 14) Potassium 40 (1460 KeV).

Figure 1: Spectre de *Xerocomus chrysenteron* (Tchécoslovaquie). Le fond est clairement visible; les éléments et les énergies correspondant aux principaux pics sont indiqués.

The origin of uranium and radium decay products in the soil:

The presence of these radionuclides is partly due to natural causes: uranium and radium may be dissolved in groundwater and migrate upwards; radon emanations have always existed and its main long-lived daughter product, Pb 210, sooner or later deposits on the soil.

These radionuclides however enter also the biosphere due to human activities:

1) Activities related to the nuclear industry:

As is the case in Issy l'Evêque, there is obviously the extraction of uranium ore, or its treatment and transformation as well as the disposal of tailings as in Gueugnon. One can also think of the production of radium for medical or industrial use.

Military tests should also be mentioned: they have released 1,4 Megacurie (1 Curie = 37 billion Becquerel) into the atmosphere between 1958 and 1962 (Robbins, 1978).

2) Activities not related to the nuclear industry:

They may also release these radioelements in the environment; all types of mining and drilling, the combustion of fossil fuels (Sheppard, 1980; Drosselmeyer,

Source : MNHN, Paris

1982) and, last but not least, agriculture: the phosphate used for fertilizers contains about 1100 to 1500 Bq/Kg of radium (Drosselmeyer, 1982).

The concentration potential of radium by the fungi:

In some cases, such as the terril de l'Héribus (Mons), the concentration factor is low. In the other cases, we had a concentration factor from 0,3 to 1. It may seem a low concentration factor and is not sufficient for a use as bio-indicator. It is nevertheless very high compared with most phanerogamous plants, which have a concentration factor of 0,1 to 0,01 (or the potential to eliminate) for radium (Sheppard, 1980; Drosselmeyer, 1982).

At first sight, the pH of the soil did not seem to be the main determining factor for the absorption; other ecological parameters are probably equally important: among those parameters, one can think of the amount of available organic matter and the specificity of the fungus metabolism. In the case of Mons (slag heap) it is plausible to think that radium present in a non-soluble form is trapped in coal particles of carboniferous shales; these particles, being of high granulometry, the radium would not be available.

The differences in absorption of radium and lead:

In some cases (in particular *C. comatus* of Gueugnon and *X. chrysenteron* of Czechoslovakia) the activity of Pb 210 measured was much higher than that of other elements of the uranium chain. In order to know whether *C. comatus* has ■ high concentration potential for lead, other results were looked for in literature and total lead was measured in the two *C. comatus* (Geugnon and Cluny) and in *C. atramentarius* (Geugnon). The results are on table 4. They clearly show that the analysed fungi do not concentrate lead. Some other explanation had to be sought:

- The radioactive lead might be present under a different (and more available) chemical form than stable lead. However, it was not possible to find a concentration factor superior to 1 anywhere in the literature.

- The mass of Pb 210 is negligible compared with the mass of stable lead: one gramme Pb 210 has an activity of 76 Ci (1 Ci = 3,7 x 10 E10 Bq). It might be that the functioning of the fungal metabolism gives different results if in presence of masses of totally different orders of magnitude. R.J. de Meijer et al. (1988) have reported a different resorption of stable cesium and radioactive cesium but this could be explained by the fact that stable cesium is much more abundant, whereas we are confronted with exactly the opposite situation here.

- There is nevertheless a third possible explanation which seems quite attractive: radon 222, the direct daughter product of Ra 226, is very soluble in water: 22,4 cm^3 radon/100 cm^3 of water at 25° (IARC study, 1988).

The mushrooms having between 80 and 93% water, they would absorb radon, in solution in water if the soil is radiferous; this radon would then decay partly or totally to Pb 210 between growth and measurement (or consumption).

This hypothesis seems quite plausible: the concentration factor lies between 0,17 and 0,09 for stable lead. It is of the same order as the results presented by Mornand (1990) and none of the other authors studied (Kuusi et al., 1981; Seeger et al., 1976; Seeger, 1982) mention *C. comatus* as being particularly lead-greedy. On the other hand the concentration of Pb 210 was sevenfold for *C. comatus* (Gueugnon) and

Source : MNHN, Paris

Sample	Soil	Fungus	Concentration factor
C. atramentarius (Gueugnon)	39,9	6,85	0,17
C. comatus (Gueugnon)	47	5,30	0,11
C. comatus (Cluny)	43	3,82	0,09

Table 4: Total lead (ppm dry matter; margin of error < 5%).
Tableau 4: Plomb total (exprimé en ppm sur la matière sèche; marge d'erreur < 5%).

also quite considerable for *X. chrysenteron*, which grew in an area far from any source of lead pollution.

The largest quantity of Pb 210, i.e. the quantity corresponding to the difference between the concentration factor for total lead (i.e. overwhelmingly stable lead) would come of the decay of radon. This makes the mushrooms unsuitable as bio-indicators - as was hoped at first - but poses a serious health problem.

Health problem and legislation:

In the European Community (but some member countries have lower limits), the annual limit for ingestion of Pb 210 is 3 000 Bq per year if it derives from a nuclear activity (Council directive N° 84/467). Obviously, legislation is not directly related to the health effect, since Pb 210 of other origins is exactly as radiotoxic as Pb 210 deriving from nuclear industry.

It must be added that this limit applies to the "added" Pb 210 only if there is no other "added" natural or artificial radioelement taken in. If there is a mixture of radionuclides, which is almost always the case, the annual limits of intake of all the other elements must be taken into account in proportion.

Thus these limits only apply if the radioactivity has its origin in the nuclear industry: it would be the case in Gueugnon (yellow cake plant) and in Issy l'Evêque (uranium mining).

Lead 210 is considered very radiotoxic because it tends to accumulate in bones: the biological half-life of lead in bones lies between 10 and 20 years (Seeger, 1982); the effective half-life (i.e. taking into account the radioactive decay) of Pb 210 in bones is of about 8 years (Drosselmeyer, 1982).

Radium is also very radiotoxic and also accumulates in bones; it has the same chemical behaviour as calcium (Sheppard, 1980). If *Coprinus comatus* of Gueugnon had been eaten - and assuming that at the time of consumption the activity of Pb 210 had been the same as at the time of measurement - the dose equivalent would have been very high: using the model of the german Institut für Strahlenhygiene (Heinrich et al., 1985), we can evaluate it as follows:

For adults, each becquerel of Pb 210 taken in delivers a dose of 2,2 x 10 E-5 Sv to the bone surface (1 sievert = 100 Rem of the former system); each becquerel of Ra 226 taken in delivers a dose of 6,8 x 10 E-6 Sv to the bone surface.

According to the same model, the effective dose to the whole body would be:

Source : MNHN, Paris

For Pb 210 (adults): 1,5 x 10 E-6 Sv for each Bq taken in;

Thus, for 722 Bq: 1,08 mSv for 1 Kg dry matter, or approximately 0,1 mSv for 1 Kg fresh matter, which means that a consumption of about 9,3 Kg is enough to reach 1mSv.

For Ra 226 (adults): 3,6 x 10 E-7 Sv for each Bq, which means that for a fungus with 136 Bq/Kg dry matter, the consumption of 1 Kg (dry) leads to a dose of 0,049 mSv, if no other radionuclide were present.

At the european level, the limit for people not involved in nuclear activities is still 5 millisievert; some countries nevertheless already follow the recommendation of the ICPR (International Radiological Protection Board), i.e. 1 mSv (100 millirem).

However, these calculations are valid only in the theoretical case in which there is no other intake of other radionuclides; in practice, there is nearly always - as here - a mixture: it must be taken into account and the limit of 1 mSv would be reached with a consumption of fungi lower than 9,3 Kg.

Determining the added radioactivity:

When dealing with sites like Gueugnon or Issy l'Evêque, knowing which proportion of radium and of its descendants is of natural or industrial origin makes it necessary to know the radiological situation before the industrial extraction or transformation began. If it is not possible, the comparison between the different elements of the family and the observation of an unbalance which cannot be explained by normal migrations of radionuclides can sometimes give ■ clue.

CONCLUSION

Many (still) unknown parameters are influencing the absorption of these radionuclides and of many other radionuclides not studied here; research based on a statistical basis may complete these first results. Provisionally, it is possible to say that none of the samples really concentrate radium: the fungi only absorb it without eliminating it; it makes them unsuitable as bio-indicators. They are not suitable either as bio-indicators for Pb 210 although some showed very high concentrations. On the other hand, the potential health problem deriving from regular consumption of fungi with levels of radioactivity as detected in some samples, in particular those growing on potentially radiferous soils and near uranium extracting or uranium transforming facilities should by no means be underestimated.

ACKNOWLEDGEMENTS

The authors wish to express their thanks to Dr. G. Kirchner, University of Bremen and to Dr. R. Courtecuisse, University of Lille for discussing the analytical techniques and reading the manuscript.

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